

## AMENDMENTS TO THE CLAIMS

This listing of claims will replace all prior versions and listings of claims in the application.

1. (currently amended) An article of manufacture suitable for use in determining whether or in what amount a chemical species is present in a target environment, which article comprises  
a multiplicity of particles in three-dimensional close-packed orientation, said particles having  
a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and  
deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and  
composed of an encapsulating monomolecular layer of ligand shell molecules that are not coupling agents, each molecule having a head-tail type structure,  
the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and  
the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, and having a tail functional group capable of selective interactions that discriminate between chemical species of interest, the tail selected from the group consisting of a heterofunctional group, an aromatic group, a secondary aliphatic group, an araliphatic group, and a tertiary aliphatic group,  
the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered; and  
a substrate suitably configured for supporting the multiplicity of particles and presenting the multiplicity of particles to contact with said environment;

wherein the particles are substantially free of coupling agents, and the multiplicity of particles is deposited by a method comprising spraying on the substrate a solution comprising the multiplicity of particles and a solvent, the substrate being at a temperature such that the solvent is flashed away or rapidly evaporated; or  
wherein the particles are linked to each other and to the substrate by coupling agents.

2. (previously presented) An article of manufacture as defined in claim 1, wherein said core comprises silver, gold, platinum or palladium, or an alloy of two or more thereof.
3. (canceled)
4. (previously presented): An article of manufacture as defined in claim 1, wherein said ligand shell molecule comprises a thiol or an amine in the head portion of the ligand shell molecule as the functional group possessing a bonding interaction with metal atoms in the core surface.
5. (canceled)
6. (previously presented) An article of manufacture as defined in claim 4, wherein said ligand shell molecule comprises an amine in the head portion.
7. (original) An article of manufacture as defined in claim 1, wherein  
in each said particle the core is of size from 2 to 20 nm in maximum dimension  
and  
the ligand shell is of thickness from 0.4 to 2.5 nm.
8. (original) An article of manufacture as defined in claim 1, wherein the particles are substantially spherical.

9. (previously presented) An article of manufacture as defined in claim 1, wherein the ligand shell molecule contains a thiol functional group in the head portion.

10-20. (canceled)

21. (currently amended) An assembly suitable for investigation of a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) a substrate suitably configured for presenting a multiplicity of particles supported thereon to contact with said environment;
  - (b) supported by said substrate, a film of from 5 to 10,000 nm comprising a multiplicity of particles in three-dimensional close-packed orientation, said particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules that are not coupling agents, each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered; and
  - (c) a sensor for monitoring said property of said multiplicity of particles;

wherein the particles are substantially free of coupling agents, and the multiplicity of particles is deposited by a method comprising spraying on the substrate a solution comprising the multiplicity of particles and a solvent, the substrate being at a temperature such that the solvent is flashed away or rapidly evaporated; or  
wherein the particles are linked to each other and to the substrate by coupling agents.

22. (previously presented) An assembly as defined in claim 21, wherein said core comprises silver, gold, platinum or palladium or an alloy of two or more thereof.

23-24. (canceled)

25. (currently amended) An assembly suitable for investigating a target environment, to determine whether or in what amount a chemical species may be present, which comprises

- (a) a substrate suitably configured for presenting a multiplicity of particles in three-dimensional close-packed orientation supported thereon to contact with said species;
- (b) supported by said substrate, said multiplicity of particles having a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules that are not coupling agents, each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and

the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, and having a tail functional group capable of selective interactions that discriminate between chemical species of interest, the tail selected from the group consisting of a heterofunctional group, an aromatic group, a secondary aliphatic group, an araliphatic group, and a tertiary aliphatic group,

the ligand shell being capable of interacting with a chemical species in a target environment such that the an electrical property of the particles is altered;

(c) a pair of electrodes, each in electrical contact with said multiplicity of particles; and

(d) a sensor for monitoring the electrical property of said multiplicity of particles to determine whether there is, or the amount of, any change in said electrical property as an indication of whether or in what amount said species is present; wherein the particles are substantially free of coupling agents, and the multiplicity of particles is deposited by a method comprising spraying on the substrate a solution comprising the multiplicity of particles and a solvent, the substrate being at a temperature such that the solvent is flashed away or rapidly evaporated; or  
wherein the particles are linked to each other and to the substrate by coupling agents.

26. (previously presented) An assembly as defined in claim 25, wherein the core comprises gold and the ligand shell molecule is selected from the group consisting of secondary aliphatic thiols, tertiary aliphatic thiols, heterofunctionally substituted aliphatic thiols, aromatic thiols, heterofunctionally substituted aromatic thiols, and heterofunctionally substituted araliphatic thiols.

27. (currently amended) A method of fabricating an assembly suitable for investigation of a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) depositing on a substrate
- (i) a pair of interdigitated electrodes each having a comb-like configuration and
  - (ii) in such manner that the electrodes are electrically connected, a thin film of a multiplicity of particles in a three-dimensional close-packed orientation having  
a core of conductive metal or conductive metal alloy, in each said particle  
the core being from 0.8 to 40.0 nm in maximum dimension and  
deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and  
composed of an encapsulating monomolecular layer of ligand shell  
molecules that are not coupling agents, each molecule having a  
head-tail type structure,  
the head being a functional group possessing a bonding interaction  
with metal atoms in the core surface, and  
the tail having a structure and composition designed to provide  
additional stabilization of metal clusters against irreversible  
agglomeration, induce solubility in solvents, and promote  
interactions with chemical species of interest, and having a  
tail functional group capable of selective interactions that  
discriminate between chemical species of interest, the tail  
selected from the group consisting of a heterofunctional  
group, an aromatic group, a secondary aliphatic group, and  
a tertiary aliphatic group,  
the ligand shell being capable of interacting with a chemical  
species in a target environment such that an electrical  
property of said multiplicity of particles is altered; and
- (b) connecting said pair of electrodes with a sensor capable of determining a change in the electrical property of said multiplicity of particles;

wherein said deposition of a thin film of the multiplicity of particles comprises spraying on the electrodes and on the substrate a solution comprising the multiplicity of particles and a solvent, said electrodes and substrate being at a temperature such that the solvent is flashed away or rapidly evaporated; or

wherein said deposition of a thin film of the multiplicity of particles comprises

(a) treating said electrodes and substrate with a difunctional material capable of binding with (i) the electrodes and the substrate and (ii) said multiplicity of said particles, such that said material binds with said electrodes and said substrate;

(b) contacting the treated electrodes and substrate with said multiplicity of particles;

(c) exposing the outwardly facing surfaces of said particles to a coupling agent capable of bonding said particles to a further multiplicity of such particles deposited thereon; and

(d) contacting the particle surfaces so exposed with said further multiplicity of particles such that:  
said further multiplicity of particles bonds with the particle surfaces, and  
the further multiplicity of particles is immobilized on those surfaces.

28. (previously presented) A method of fabrication as defined in claim 27, wherein said deposition of a thin film of the multiplicity of particles comprises spraying on the electrodes and on the substrate a solution comprising the multiplicity of particles and a solvent, said electrodes and substrate being at a temperature such that the solvent is flashed away or rapidly evaporated.

29. (previously presented) A method of fabrication as defined in claim 27, wherein said deposition of a thin film of the multiplicity of particles comprises
- (a) treating said electrodes and substrate with a difunctional material capable of binding with (i) the electrodes and the substrate and (ii) said multiplicity of said particles, such that said material binds with said electrodes and said substrate;
  - (b) contacting the treated electrodes and substrate with said multiplicity of particles;
  - (c) exposing the outwardly facing surfaces of said particles to a coupling agent capable of bonding said particles to a further multiplicity of such particles deposited thereon; and
  - (d) contacting the particle surfaces so exposed with said further multiplicity of particles such that:  
said further multiplicity of particles bonds with the particle surfaces, and  
the further multiplicity of particles is immobilized on those surfaces.
- 30-31. (canceled)
32. (previously presented) A method of fabrication as defined in claim 29, wherein said cycle is performed a plurality of times.
33. (currently amended) A system suitable for investigating a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) a multiplicity of particles in three-dimensional close-packed orientation, said particles having  
a core of conductive metal or conductive metal alloy, in each said particle such  
core being of 0.8 to 40.0 nm in maximum dimension and  
deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and  
composed of an encapsulating monomolecular layer of ligand shell  
molecules that are not coupling agents, each molecule having a head-tail  
type structure,

the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and  
the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest,  
the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered;

- (b) means for exposing said multiplicity of particles to said environment, said means for exposing having a substrate suitably configured for supporting the multiplicity of particles;
- (c) means for subjecting said multiplicity of particles to conditions sufficient for said property to be exhibited; and
- (d) means for monitoring said property to determine whether there is, or the amount of, any change in said property as an indication of whether or in what amount said species is present;

wherein the particles are substantially free of coupling agents, and the multiplicity of particles is deposited by a method comprising spraying on the substrate a solution comprising the multiplicity of particles and a solvent, the substrate being at a temperature such that the solvent is flashed away or rapidly evaporated; or  
wherein the particles are linked to each other and to the substrate by coupling agents.

34. (currently amended) A system for investigating a target environment to determine whether or in what amount a chemical species may be present, which comprises
- (a) a multiplicity of particles in three dimensional close-packed orientation, said particles having  
a core of conductive metal or conductive metal alloy, in each said particle such core being of 0.8 to 40.0 nm in maximum dimension and

deposited on said core a ligand shell of thickness from 0.4 to 4.0 nm and composed of an encapsulating monomolecular layer of ligand shell molecules that are not coupling agents, each molecule having a head-tail type structure, the head being a functional group possessing a bonding interaction with metal atoms in the core surface, and the tail having a structure and composition designed to provide additional stabilization of metal clusters against irreversible agglomeration, induce solubility in solvents, and promote interactions with chemical species of interest, the ligand shell being capable of interacting with a chemical species in a target environment such that an electrical property of said multiplicity of particles is altered;

- (b) means for exposing said multiplicity of particles to said environment, said means for exposing having a substrate suitably configured for supporting the multiplicity of particles;
- (c) means for passing an electrical field through said multiplicity of particles; and
- (d) means for monitoring the electrical property of said multiplicity of particles to determine whether there is, or the amount of, any change in said electrical property as an indication of whether or in what amount said species is present; wherein the particles are substantially free of coupling agents, and the multiplicity of particles is deposited by a method comprising spraying on the substrate a solution comprising the multiplicity of particles and a solvent, the substrate being at a temperature such that the solvent is flashed away or rapidly evaporated; or wherein the particles are linked to each other and to the substrate by coupling agents.

35. (original) A system as defined in claim 34, wherein said means for monitoring the electrical resistivity of said multiplicity of particles includes a current-to-voltage converter circuit followed by a precision rectifier and low-pass filter.

36. (original) A system as defined in claim 35, wherein said means further includes a voltage-to-frequency converter.
37. (previously presented) An article of manufacture as defined in claim 1, wherein said heterofunctional group is OH, COOH, or Cl.
38. (previously presented) An article of manufacture as defined in claim 1 further comprising a ligand molecule having the formula  $\text{HS}(\text{CH}_2)_6\text{OH}$ .
39. (canceled)
40. (previously presented) An article of manufacture as defined in claim 1, wherein the ligand shell molecule contains a thiol functional group in the head portion of the structure and the heterofunctional group comprising individually or a combination of an alcohol, phenol, fluoroalcohol, carboxylic acid, ether, phosphoryl, or halide heteroatom functional groups.
41. (previously presented) An article of manufacture as defined in claim 1, wherein said ligand shell molecule comprises a thiol in the head portion of the structure and comprises a secondary or tertiary aliphatic hydrocarbon structure, an aromatic hydrocarbon structure, an araliphatic hydrocarbon structure, a heterofunctional aliphatic structure, a heterofunctional aromatic structure, or a heterofunctional araliphatic structure in the tail portion of the ligand shell molecule structure.
42. (previously presented) An article of manufacture as defined in claim 41, wherein said heterofunctionality comprises a hexafluoroacetone derived adduct.
43. (previously presented) A method of fabrication as defined in claim 29, wherein said difunctional material comprises dithiol and silane thiol coupling agents.

44. (currently amended) A method of fabrication as defined in claim 29 ~~claim 31~~, wherein said coupling agent comprises a dithiol molecule.
45. (previously presented) A method of fabrication as defined in claim 29, wherein said difunctional material comprises a dithiol molecule.
46. (canceled)
47. (previously presented) An article of manufacture as defined in claim 1, wherein said ligand shell molecule is selected from the group consisting of benzyl mercaptan, phenylethyl mercaptan, araliphatic mercaptan, and 4-methoxybenzyl mercaptan.
48. (currently amended) A method of fabrication as defined in claim 29 ~~claim 31~~, wherein said coupling agent is selected from the group consisting of dithiol, octanedithiol, and hexanedithiol.
49. (previously presented) An assembly as defined in claim 21, wherein the film is up to about 400 nm thick.
50. (previously presented) An article of manufacture as defined in claim 1, wherein said aromatic group, said secondary aliphatic group, or said tertiary aliphatic group is substituted by OH, COOH, or Cl.
51. (previously presented) An article of manufacture as defined in claim 1, wherein the tail functional group is selected from the group consisting of heterofunctional group and aromatic group.
52. (canceled)
53. (currently amended) The article of manufacture ~~assembly~~ of claim 1, wherein the particles are linked to each other and to the substrate by coupling agents.

54. (previously presented) The method of claim 27, wherein the particles are substantially free of coupling agents.
55. (previously presented) The method of claim 27, wherein the particles are linked to each other and to the substrate by coupling agents.
56. (currently amended) The article of claim 1 ~~claim 52~~, wherein the particles are substantially free of coupling agents and the multiplicity of particles is deposited by a method comprising spraying on the substrate a solution comprising the multiplicity of particles and a solvent, the substrate being at a temperature such that the solvent is flashed away or rapidly evaporated.
57. (previously presented) The method of claim 28, wherein the particles are substantially free of coupling agents.